PATENT ABSTRACTS OF JAPAN

(11)Publication number : 2001-046864

(43)Date of publication of application : 20.02.2001

(51)Int.Cl. B01J 8/02

B01J 19/00

C01B 31/02

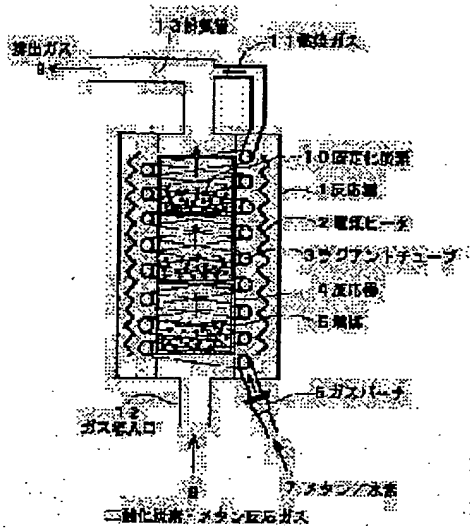
(21)Application number : 11-223172

(22)Date of filing : 06.08.1999

(71)Applicant : SHIMADZU CORP
RESEARCH INSTITUTE OF INNOVATIVE TECHNOLOGY FOR THE EARTH

(72)Inventor : TANAKA SHUICHI
YAMAMOTO MASANOBU

(54) CARBON DIOXIDE FIXING APPARATUS



(57)Abstract:

PROBLEM TO BE SOLVED: To conserve heat energy as the heating source of a reaction tank.

SOLUTION: A radiant tube 3 is wound around the outer periphery of a reactor 4 housing a catalyst 5 and methane being a part of reaction gas or hydrogen capable of decomposing methane is burnt by a gas burner 6 and the combustion gas 11 of the gas burner is sent to the radiant tube 3 to be used as a heating source. Carbon dioxide/methane reaction gas 8 is introduced into the reaction tank 1 from a gas introducing port 12 provided to the lower part of the reaction tank 1 and fixed carbon 10 is formed on the surface of the catalyst 5 by catalytic reaction. The gas after reaction and the combustion gas 11 are discharged to the outside from an exhaust pipe 13 provided to the upper part of the reaction tank 1 to be circulated. If reaction is stabilized, only an electric heater is used in heat insulation by little power.

[Claim(s)]

[Claim 1]A carbon dioxide fixing apparatus which fixes carbon dioxide by making into a source of reactant gas carbon dioxide and methane having the following, passing a heat flow rate of the combustion part to said radiant tube, and controlling temperature of said reactor.

A radiant tube for heating spirally piped by periphery of a reactor.

A combustion part which burns hydrogen gas which disassembles and obtains methane or methane.

[Claim 2]A carbon dioxide fixing apparatus of claim 1 forming an electric heater the outside of the above-mentioned radiant tube, or by turns.

[Detailed Description of the Invention]

[0001]

[Field of the Invention]This invention relates to the temperature control of the reactor for advancing catalytic reaction especially with respect to the carbon dioxide fixing apparatus which makes carbon dioxide and methane a gas source.

[0002]

[Description of the Prior Art]In order to reduce the greenhouse gas concentration in the atmosphere, it is indispensable to suppress the discharge itself. Methods various for the discharge reduction are studied. To one of the methods of fixing and recycling CO_2 discharged in large quantities by a discharge source from the carbon dioxide (CO_2) in the atmosphere, plant, an iron mill, a cement plant, etc. For example, CO_2 is returned under hydrogen (H_2) atmosphere and the method of changing into fine-powder-form carbon is devised. The C0 2 decollator with which the conversion method separates CO_2 from the atmosphere or exhaust gas, It comprises a CO_2 concentration device which condenses the separated CO_2, a CO_2/H_2 reaction apparatus which makes CO_2 and H_2 react under catalyst existence, and generates fine-powder-form carbon, etc. A distribution diagram until drawing 2 is fixed from a carbon dioxide source of release is shown. The gas emitted from the CO_2 sources of release 14, such as a factory which burns the fossil fuel 15, is incorporated into the CO_2 decollator 16, only carbon dioxide is separated, and it introduces into CO_2 concentration device 17. It is mixed with hydrogen from the outside, or methane, and the carbon dioxide condensed with CO_2 concentration device 17 is introduced into the CO_2 fixed device 18. Gas pressure is increased by the compressor 18a, and the introduced reactant gas is sent to the heat exchanger 18b which exchanges the gas heat of the return after the reaction of the reaction vessel 18c. The warmed reactant gas goes into the reaction vessel 18c, 18 d of catalysts decompose and fix it, and the fixed carbon adheres to the surface of 18 d of catalysts. Hydrogen of a surplus with a high temperature generated in reaction time, a steam, unreacted carbon dioxide, and methane are discharged from the reaction vessel 18c. Heat exchange of the gas is carried out by the heat exchanger 18b, it goes into the condenser 18e and is cooled, and a steam is condensed, serves as water and is discharged outside. The remaining gas can raise a pressure by the compressor 18a again, and is sent out to a circulation circuit. The catalyst/carbon fixed by the reaction vessel 18a are taken out from 18h of catalyst flow tub maintaining structure of the lower part of the reaction vessel 18c, the fixed carbon which is a catalyst / carbon eliminator 18g, and adhered to 18 d of catalysts and 18d of catalyst surface is separated, and carbon is taken out outside. It is reproduced with the catalyst regeneration machine 18f, and 18 d of catalysts are again supplied to the reaction vessel 18c.

[0003]The carbon dioxide fixing apparatus which makes CO_2 and H_2 which decomposes and can do CH_4 react under catalyst existence, and generates fine-powder-form carbon is shown in drawing 3. Inside the reaction vessel 20, the reactor 4 is formed in two or more steps, and the catalyst 5 is placed into the reactor 4, and the far infrared heater 19 is set to the heating furnace 21 for promoting catalytic reaction by the outside, and it

works as a heater for heating on it. Carbon dioxide and the methane response gas 8 are introduced from the gas inlet 12 established in the lower part of the reaction vessel 9, and the reaction vessel 20 is heated with the far infrared heater 19 at the time of reaction starting. The temperature of the catalyst 5 rises and catalytic reaction decomposes methane into carbon and hydrogen. And carbon dioxide reacts to the hydrogen and becomes carbon and water (steam). Carbon is fixed by the surface of the catalyst 5 as the fixed carbon 10, and comes out outside from the exhaust pipe 22 with which the gas of a steam, excessive hydrogen, and unreacted methane and unreacted carbon dioxide was formed in the central part of the reaction vessel 20 as the emission gas 9.

[0004]

[Problem(s) to be Solved by the Invention]Although the conventional carbon dioxide fixing apparatus is constituted as mentioned above, if methane of carbon dioxide and the methane response gas 8 does not carry out the endoergic reaction of CH_4=C+2H $_2$+90.1 kj/mol first according to the catalyst 5, hydrogen does not occur. If hydrogen does not exist, fixed reaction:CO_2+2H $_2$=C+2H $_2$O-96 kj/mol of carbon dioxide is not performed. Therefore, the heating furnace 21 is established in the reaction vessel 20, and temperature up of the temperature of the reactor 4 is carried out with the far infrared heater 19 by the electrical and electric equipment as a source of heating. While the reaction is advancing uniformly, the whole reaction must be heated for the heat loss to the circumference, although what is necessary is just to become CH_4+CO_2=2C+2H $_2$O-5.9 kj/mol and to apply a little heat from the outside. Since a reaction vessel becomes large and a reaction vessel is heated by 500-700 ** as a device is put in practical use, heating power becomes large too much only by an electric heating. The more it comes to perform carbon-dioxide-fixation-ization on a large scale, the more SUBJECT that thermal energy required for the employment must be saved labor occurs.

[0005]This invention was made in view of such a situation, and is ****. The purpose is to provide the carbon dioxide fixing apparatus which used the gas of the carbon dioxide fixing apparatus effectively, and attained laborsaving of thermal energy as a source of heating of 20.

[0006]

[Means for Solving the Problem]In order to attain the above-mentioned purpose, a carbon dioxide fixing apparatus of this invention, In a carbon dioxide fixing apparatus which fixes carbon dioxide by making carbon dioxide and methane into a source of reactant gas, A periphery of a reactor is equipped with a combustion part which burns

hydrogen gas which disassembles and obtains a radiant tube, and methane or methane for heating piped spirally, a heat flow rate of the combustion part is passed to said radiant tube, and temperature of said reactor is controlled.

[0007]An electric heater is formed the outside of the above-mentioned radiant tube, or by turns.

[0008]A carbon dioxide fixing apparatus of this invention is constituted as mentioned above, and hydrogen gas which disassembles and obtains methane or methane used for this device is burned, Since it lets a heat flow rate pass to a radiant tube for heating spirally piped by periphery of a reactor, catalytic reaction of carbon dioxide can be performed to it only in this heat source, and energy can be used effectively for it. It can also carry out by attaching an electric heater in addition to a gas heating method.

[0009]

[Embodiment of the Invention]One example of the carbon dioxide fixing apparatus of this invention is described referring to drawing 1. The gas inlet 12 where this device introduces the reaction vessel 1, and the carbon dioxide and methane response gas 8 which were formed in the lower part, It comprises the gas burner 6 which sends combustion gas into the radiant tube 3 provided in the reaction vessel 1, and the exhaust pipe 13 which discharges the gas and the combustion gas 11 after the reaction provided in the upper part. The reactor 4 is set to an inside by two or more steps, the reaction vessel 1 is put into the catalyst 5 inside the reactor 4, and the radiant tube 3 is spirally rolled on the circumference of the exterior. And the electric heater 2 is formed the exterior or the radiant tube 3, and by turns. The radiant tube 3 is connected to the gas burner 6 provided in the lower part of the reaction vessel 1. The hydrogen gas which can decompose and do the methane supplied as reactant gas is used, the hydrogen gas is burned, and hot combustion gas is sent into the radiant tube 3. With the heat from the radiant tube 3, carbon dioxide and the methane response gas 8 react to the catalyst 5, and the fixed carbon 10 generates on the surface of the catalyst 5. nickel in which the catalyst 5 makes SiO_2 and aluminum$_2O_3$ a carrier, for example, Co, etc. are used. Methane of the reactant gas supplied is used for the methane burned with the gas burner 6, and the hydrogen generated by the reaction vessel 1 in the following processes is used for hydrogen. If carbon dioxide and the methane response gas 8 are introduced from the lower part of the reaction vessel 1 and the reactor 4 is heated with the electric heater 2, the temperature of the catalyst 5 will rise and methane will be disassembled into carbon and hydrogen. And carbon dioxide reacts to the hydrogen and becomes carbon and water (steam). Carbon is fixed by the catalyst surface and the gas of a steam, excessive hydrogen, and unreacted methane and unreacted carbon dioxide is discharged

from the exhaust pipe 13 of the upper part of the reaction vessel 1. Externally, hydrogen and other gas (a steam, carbon dioxide, methane) are separated, hydrogen is taken out, and the gas burner 6 is supplied and is burned. And at the time of the temperature up which carries out temperature up of the reactor 4 with the radiant tube 3, if heating by gas is performed and it becomes a predetermined temperature, it will be kept warm only with the electric heater 2, and catalytic reaction will be advanced.

[0010]Next, the process of carbon-dioxide-fixation-izing of this device is explained. First, the catalyst 5 is put on each reactor 4. And if methane / hydrogen gas 7 is burned with the gas burner 6 and combustion gas is passed to the radiant tube 3, the radiant tube 3 will be heated and the temperature of the reactor 4 containing the catalyst 5 will be heated by 500-700 **. Next, from the gas inlet 12 established in the lower part of the reaction vessel 1, if carbon dioxide and the methane response gas 8 are introduced, First, CH_4 gas of carbon dioxide and the methane response gas 8 carries out the reaction of $CH_4=C+2H_2+90.1$ kJ/mol according to the catalyst 5 (that with which nickel [which makes SiO_2 and $aluminum_2O_3$ a carrier], Co, etc. were filled up). And CO_2 gas of carbon dioxide and the methane response gas 8 carries out the reaction of $CO_2+2H_2=C+2H_2O-96$ kJ/mol according to the catalyst 5 (that with which nickel [which makes SiO_2 and $aluminum_2O_3$ a carrier], Co, etc. were filled up). Therefore, the whole reaction serves as $CH_4+CO_2=2C+2H_2O-5.9$ kJ/mol. Since the temperature of reaction time should just apply a little heat from the outside while catalytic reaction is progressing at 500-700 **, it is controlled with accuracy sufficient to a required temperature by heating of the radiant tube 3 by the gas combustion of the gas burner 6, or the electric heater 2. And carbon adheres to the surface of the catalyst 5 and carbon dioxide and the methane response gas 8 are fixed by catalytic reaction as the fixed carbon 10. The gas discharged from the exhaust pipe 13 of the upper part of the reaction vessel 1, Hydrogen is used as an object for combustion, it is cooled with a condenser, and a steam becomes water, and is taken out outside, it is gas of a steam, excessive hydrogen, and unreacted methane and unreacted carbon dioxide, and it is used [the gas of unreacted methane and unreacted carbon dioxide is again returned to a circulation circuit, and] as reactant gas.

[0011]In the process in which carbon dioxide reacts to hydrogen and it becomes carbon and water with the above-mentioned reaction formula, methane reacts according to a catalyst, the hydrogen quantity consumed decomposes, and if it becomes less than the yield of hydrogen of the process in which it becomes carbon and hydrogen, excessive hydrogen will occur. Hydrogen of the surplus generated in the above-mentioned reaction and $CH_4=C+2H_2+90.1$ kJ/mol is taken out outside, and this hydrogen is used as combustion gas of the gas burner 6.

[0012]The more it comes to perform carbon-dioxide-fixation-ization on a large scale, the more thermal energy required for the employment must be saved labor. Therefore, the hydrogen obtained by disassembly of the methane used for a reaction or methane is used for this device, it is burned with the gas burner 6, is making the radiant tube 3 the periphery of the reactor 4 with the heat source, and becomes effective use of energy compared with the electric heater heating method by the conventional electric power.

[0013]

[Effect of the Invention]The carbon dioxide fixing apparatus of this invention is constituted as mentioned above, and a reaction vessel becomes large as a device is put in practical use. Since a reaction vessel is heated by 500-700 **, heating power becomes large too much only by an electric heating. With this device, while heating by gas is performed and catalytic reaction is advancing at a predetermined temperature, at the time of temperature up, since little thermal energy may be used, if it is kept warm with an electric heater, it will become energy saving, and a running cost reduces it, and it can perform highly precise temperature control.

[Field of the Invention]This invention relates to the temperature control of the reactor for advancing catalytic reaction especially with respect to the carbon dioxide fixing apparatus which makes carbon dioxide and methane a gas source.

[Description of the Prior Art]In order to reduce the greenhouse gas concentration in the atmosphere, it is indispensable to suppress the discharge itself. Methods various for the discharge reduction are studied. To one of the methods of fixing and recycling CO_2 discharged in large quantities by a discharge source from the carbon dioxide (CO_2) in the atmosphere, plant, an iron mill, a cement plant, etc. For example, CO_2 is returned under hydrogen (H_2) atmosphere and the method of changing into fine-powder-form carbon is devised. The $C0_2$ decollator with which the conversion method separates CO_2 from the atmosphere or exhaust gas, It comprises a CO_2 concentration device which condenses the separated CO_2, a CO_2/H_2 reaction apparatus which makes CO_2 and H_2 react under catalyst existence, and generates fine-powder-form carbon, etc. A distribution diagram until drawing 2 is fixed from a carbon dioxide source of release is shown. The gas emitted from the CO_2 sources of release 14, such as a factory which burns the fossil fuel 15, is incorporated into the CO_2 decollator 16, only carbon dioxide is separated, and it introduces into CO_2 concentration device 17. It is mixed with hydrogen from the outside, or methane, and the carbon dioxide condensed with CO_2 concentration device 17 is introduced into the CO_2 fixed device 18. Gas pressure is increased by the compressor 18a, and the introduced reactant gas is sent to the heat exchanger 18b which exchanges the gas heat of the return after the reaction of the reaction vessel 18c.

The warmed reactant gas goes into the reaction vessel 18c, 18 d of catalysts decompose and fix it, and the fixed carbon adheres to the surface of 18 d of catalysts. Hydrogen of a surplus with a high temperature generated in reaction time, a steam, unreacted carbon dioxide, and methane are discharged from the reaction vessel 18c. Heat exchange of the gas is carried out by the heat exchanger 18b, it goes into the condenser 18e and is cooled, and a steam is condensed, serves as water and is discharged outside. The remaining gas can raise a pressure by the compressor 18a again, and is sent out to a circulation circuit. The catalyst/carbon fixed by the reaction vessel 18a are taken out from 18h of catalyst flow tub maintaining structure of the lower part of the reaction vessel 18c, the fixed carbon which is a catalyst / carbon eliminator 18g, and adhered to 18 d of catalysts and 18d of catalyst surface is separated, and carbon is taken out outside. It is reproduced with the catalyst regeneration machine 18f, and 18 d of catalysts are again supplied to the reaction vessel 18c.

[0003]The carbon dioxide fixing apparatus which makes CO_2 and H_2 which decomposes and can do CH_4 react under catalyst existence, and generates fine-powder-form carbon is shown in drawing 3. Inside the reaction vessel 20, the reactor 4 is formed in two or more steps, and the catalyst 5 is placed into the reactor 4, and the far infrared heater 19 is set to the heating furnace 21 for promoting catalytic reaction by the outside, and it works as a heater for heating on it. Carbon dioxide and the methane response gas 8 are introduced from the gas inlet 12 established in the lower part of the reaction vessel 9, and the reaction vessel 20 is heated with the far infrared heater 19 at the time of reaction starting. The temperature of the catalyst 5 rises and catalytic reaction decomposes methane into carbon and hydrogen. And carbon dioxide reacts to the hydrogen and becomes carbon and water (steam). Carbon is fixed by the surface of the catalyst 5 as the fixed carbon 10, and comes out outside from the exhaust pipe 22 with which the gas of a steam, excessive hydrogen, and unreacted methane and unreacted carbon dioxide was formed in the central part of the reaction vessel 20 as the emission gas 9.